Influences of hydroxyl radicals (OH) on top-down estimates of the global and regional methane budgets

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Abstract
The hydroxyl radical (OH), which is the dominant sink of methane (CH₄), plays a key role to close the global methane budget. Previous research that assessed the impact of OH changes on the CH₄ budget mostly relied on box modeling inversions with a very simplified atmospheric transport and no representation of the heterogeneous spatial distribution of OH radicals. Here using a variational Bayesian inversion framework and a 3D chemical transport model, LMDz, combined with 10 different OH fields derived from chemistry-climate models (CCMI experiment), we evaluate the influence of OH burden, spatial distribution, and temporal variations on the global CH₄ budget. The global tropospheric mean CH₄-reaction-weighted [OH] ([OH]_{GM, CH₄}) ranges 10.3-16.3×10^5 molec cm⁻³ across 10 OH fields during the early 2000s, resulting in inversion-based global CH₄ emissions between 518 and 757 Tg yr⁻¹. The uncertainties in CH₄ inversions induced by the different OH fields are comparable to, or even larger than the uncertainty typically given by bottom-up and top-down estimates. Based on the LMDz inversions, we estimate that a 1%-increase in OH burden leads to an increase of 4 Tg yr⁻¹ in the estimate of global methane emissions, which is about 25% smaller than what is estimated by box-models. The uncertainties in emissions induced by OH are largest over South America, corresponding to large inter-model differences of [OH] in this region. From the early to the late 2000s, the optimized CH₄ emissions increased by 21.9±5.7 Tg yr⁻¹ (16.6-30.0 Tg yr⁻¹), of which ~25% (on average) is contributed by -0.5 to +1.8% increase in OH burden. If the CCMI models represent the OH trend properly over the 2000s, our results show that a higher increasing trend of CH₄ emissions is needed to match the CH₄ observations compared to the CH₄ emission trend derived using constant OH. This study strengthens the importance to reach a better representation of OH burden and of OH spatial and temporal distributions to reduce the uncertainties on the global CH₄ budget.
1 Introduction

Methane (CH$_4$) plays an important role in both climate change and air quality as a major greenhouse gas and tropospheric ozone precursor (Ciais et al., 2013). CH$_4$ is emitted from various anthropogenic sources including agriculture, waste, fossil fuel, and biomass burning, as well as natural sources including wetlands and other freshwater systems, geological sources, termites, and wild animals. CH$_4$ is removed from the atmosphere mainly by reaction with hydroxyl radical (OH) (Saunois et al. 2016, 2017). Tropospheric CH$_4$ levels have more than doubled between the 1850s and present-day (Etheridge et al., 1998) in response to anthropogenic emissions and climate variabilities, leading to about 0.62 W m$^{-2}$ additional radiative forcing (Etminan et al., 2016) and increases in tropospheric ozone levels of ~5 ppbv (Fiore et al., 2008). The global CH$_4$ atmospheric mixing ratio stabilized in the early 2000s but resumed growing at a rate of ~5 ppbv yr$^{-1}$ since 2007 (Dlugokencky, NOAA/ESRL, 2019).

Explaining the CH$_4$ stabilization and renewed growth requires an accurate estimation of the CH$_4$ budget and its evolution, as the source-sink imbalance that is responsible for the contemporary CH$_4$ yearly growth only accounts for 3% of the total CH$_4$ burden (Turner et al., 2019). To reconcile the uncertainties in the current estimation of CH$_4$ emissions from various sources, the Global Carbon Project integrates top-down and bottom-up studies and approaches (Kirschke et al. 2013; Saunois et al., 2016; 2017; 2019). However, gaps still remain in global and regional CH$_4$ emissions estimated by top-down and bottom-up approaches, as well as within each approach (Kirschke et al. 2013; Saunois et al., 2016; Bloom et al., 2017). The top-down method, which optimizes emissions by assimilating observations in an atmospheric inversion system, is expected to reduce uncertainties of bottom-up estimates. Among the remaining causes of uncertainties in the global methane budget, the representation of CH$_4$ sinks, mainly from OH oxidation, is one of the largest, as seen by process-based models for atmospheric chemistry (Saunois et al., 2017).

OH is the most important tropospheric oxidizing agent determining the lifetime of many pollutants and
greenhouse gases including CH$_4$ (Levy, 1971). A small perturbation of OH can result in significant changes in atmospheric CH$_4$ through catalytic chemistry (Turner et al., 2019). Tropospheric OH is mainly produced by the reaction of excited oxygen atoms (O($^1$D)) with water vapor (primary production) and the reaction of nitrogen oxide (NO) with hydroperoxyl radicals (HO$_2$) and organic peroxy radicals (RO$_2$) (secondary production). OH is rapidly removed by carbon monoxide (CO), methane (CH$_4$), and non-methane volatile organic compounds (NMVOCs) (Logan et al., 1981; Lelieveld et al., 2004). Tropospheric OH has a very short lifetime of a few seconds (Logan et al., 1981; Lelieveld et al., 2004), hindering direct measurements and estimates of global OH concentrations ([OH]) and limits our current understanding of the CH$_4$ sink.

Global tropospheric [OH] is approximately $1 \times 10^6$ molec cm$^{-3}$ as calculated by atmospheric chemistry models (Naik et al., 2013; Voulgarakis et al., 2013, Zhao et al., 2019) and inversions of 1,1,1-trichloroethane (methyl chloroform, MCF) (Prinn et al., 2001; Bousquet et al., 2005; Montzka et al., 2011), resulting in a chemical lifetime of ~9 years for tropospheric CH$_4$ (Naik et al., 2013). However, accurate estimation of [OH] magnitude, distributions, and year-to-year variations needed for CH$_4$ emission optimizations are still evolving. For global tropospheric [OH], both MCF inversions and atmospheric chemistry model inter-comparisons give a 10%-15% uncertainty (Prinn et al., 2001; Bousquet et al., 2005; Naik et al., 2013; Zhao et al., 2019). For [OH] spatial distributions, MCF-based inversions generally infer similar mean [OH] over both hemispheres (Bousquet et al., 2005; Patra et al., 2014), while atmospheric chemistry models generally give [OH] Northern hemisphere to Southern hemisphere (N/S) ratios above 1 (e.g. Naik et al., 2013; Zhao et al., 2019). For [OH] year-to-year variations, some studies have estimated magnitudes significant enough to help explain part the stagnation in atmospheric CH$_4$ concentrations during the early 2000s (Rigby et al., 2008; McNorton et al., 2016; Dalsøren et al., 2016; Rigby et al., 2017; Turner et al., 2017), whereas others show smaller trends and inter-annual variations for [OH] (Montzka et al, 2011; Naik et al., 2013; Voulgarakis et al., 2013; Zhao et
In a recent study, Zhao et al. (2019) simulated CH$_4$ with an ensemble of OH fields and showed that uncertainties in [OH] variations could explain up to 54% of model-observation discrepancies in surface CH$_4$ mixing ratio changes from 2000 to 2016.

Current top-down estimates of the global CH$_4$ budget usually prescribe constant [OH] over time and attribute model-observation mismatches exclusively to CH$_4$ emissions (Saunois et al., 2017). The role of OH on the top-down estimation of CH$_4$ emissions has been mostly evaluated using two box-model inversions (e.g. Rigby et al., 2017; Turner et al., 2017). Such a simplified representation of atmospheric processes, however, does not consider spatial heterogeneities in atmospheric transport, emission distributions, and OH vertical and horizontal distributions (Naus et al., 2019). Following the earlier work of Zhao et al. (2019), here we use the LMDZ-SACS (Laboratoire de Météorologie Dynamique model with Zooming capability-Simplified Atmospheric Chemistry System) 3D chemical transport model forced with 10 different OH fields derived from chemistry-climate models (Patra et al., 2011; Szopa et al., 2013; Hegglin and Lamarque, 2015; Morgenstern et al., 2017; Zhao et al., 2019; Terrenoire et al., 2019) to evaluate the influence of OH distribution and trends on the global CH$_4$ budget as estimated by a Bayesian 4D inversion system. Section 2 briefly describes the OH fields, the inversion method, and the setups of inversion experiments. Section 3 illustrates the influence of OH on the top-down estimation of CH$_4$ budgets and variations, specifically: i) global, regional, and sectoral CH$_4$ emissions (Section 3.1), ii) emission changes between the early 2000s and late 2000s (Section 3.2), and iii) year-to-year variations in methane emissions (Section 3.3). Section 4 summarizes the results and discusses the impact of OH on the current CH$_4$ budget.

2 Method

2.1 OH fields

In this study, we test the 10 OH fields presented in by Zhao et al. (2019), including 7 OH fields simulated
by chemistry-transport and chemistry-climate models from Phase 1 of the Chemistry-Climate Model Initiative (CCMI) (Hegglin and Lamarque, 2015; Morgenstern et al., 2017), 2 OH fields simulated by the Interaction with Chemistry and Aerosols (INCA) model coupled to the general circulation model of the Laboratoire de Météorologie Dynamique (LMD) model (Hauglustaine et al., 2004; Szopa et al., 2013), and 1 OH field from the TRANSCOM-2011 inter-comparison exercise (Patra et al., 2011) (Table 1).

The CCMI project conducted simulations with 20 state-of-the-art atmospheric chemistry-climate and chemistry-transport models to evaluate the model’s projections of atmospheric composition (Hegglin and Lamarque, 2015; Morgenstern et al., 2017). In order to force atmospheric inversions during 2000-2010, we use OH fields from 7 of the 20 CCMI model simulations of REF-C1 experiments (Table 1), which were driven by observed sea surface temperatures and state-of-the-art historical forcings (covering 1960-2010). For the inversions after 2010 (only with the CESM1-WACCM model, see Section 2.3), we apply inter-annual variations of OH generated from REF-C2 experiments, which were driven by sea surface conditions calculated by the online-coupled ocean and sea ice modules. Although all of the CCMI models use the same anthropogenic emission inventories, the simulated OH fields show different spatial and vertical distributions, as well as year-to-year variations (Zhao et al., 2019). The inter-model differences of OH are mainly attributable to differences in natural emissions (primarily biogenic VOCs and soil and lightning NOx emissions) and in chemical mechanisms related to NO production and loss (Zhao et al., 2019), as well as the atmospheric transport of the chemical species in the models.

The two INCA OH fields, INCA NMHC-AER-S and INCA NMHC are simulated by two different versions of the INCA (Interaction with Chemistry and Aerosols) chemical model coupled to LMDz (Szopa et al., 2013; Terrenoire et al., 2019). The main difference between the two simulations is that INCA NMHC-AER-S includes both gas-phase and aerosol chemistry in the whole atmosphere while INCA NMHC only includes gas-phase chemistry in the troposphere (Szopa et al., 2013; Terrenoire et al., 2019).
We also include the climatological OH field used in the TransCom simulations (Patra et al., 2011), which in the troposphere uses the semi-empirical, observation-based OH field computed by Spivakovsky et al. (2000).

Table 1 summarizes the global tropospheric mean CH$_4$-reaction-weighted [OH] ([OH]$_{GM}$-CH$_4$, [OH] weighted by reaction rate of OH with CH$_4$ ($K_{OH+CH4} \times$ dry air mass, Lawrence et al., 2001) and dry air mass-weighted [OH] ([OH]$_{GM-M}$), as well as inter-hemispheric ratios (N/S ratios) calculated with [OH]$_{GM}$-CH$_4$ for the 10 OH fields used in this study. The [OH]$_{GM}$-CH$_4$ is a better indicator of the global atmospheric oxidizing efficiency for CH$_4$ than [OH]$_{GM-M}$, since the latter is insensitive to the CH$_4$+OH reaction rate increased significantly with temperature (Lawrence et al., 2001). Both the mean value ($12.3 \pm 3.8 \times 10^5$ molec cm$^{-3}$) and absolute range ($10.3-16.3 \times 10^5$ molec cm$^{-3}$) of [OH]$_{GM}$-CH$_4$ calculated for the 10 OH fields are larger than those of [OH]$_{GM-M}$ ($11.4 \pm 2.8 \times 10^5$ molec cm$^{-3}$ and $9.4-14.4 \times 10^5$ molec cm$^{-3}$, respectively), as some of the OH fields show distinct vertical distributions (Zhao et al., 2019). The inter-hemispheric OH ratios range from 1.0 to 1.5, larger than ones derived from MCF inversions (e.g. Bousquet et al., 2005; Patra et al., 2014). A comprehensive analysis of spatial and vertical distributions of these OH fields was presented in Zhao et al. (2019).

2.2 Inverse method

We conduct an ensemble of variational inversions of CH$_4$ budget that rely on Bayes’ theorem (Chevallier et al., 2005) with the same set of atmospheric observations of CH$_4$ mixing ratios but different prescribed OH fields as described in Sect. 2.1. A variational data assimilation system optimizes CH$_4$ emissions and sinks by minimizing the cost function $J$, defined as:

$$J(x) = \frac{1}{2} (x - x^b)^T B^{-1} (x - x^b) + \frac{1}{2} (H(x) - y)^T R^{-1} (H(x) - y)$$  

(1)

where $x$ is the control vector that includes total CH$_4$ emissions per 10 days at the model resolution of
3.75° (in longitude) × 1.85° (in latitude) and initial conditions at longitudinal and latitudinal bands of 20° × 15°; $x^b$ is the prior of the control vector $x$; $y$ is the observation vector of observed CH$_4$ mixing ratios, here at the surface; and $H(x)$ represents the sensitivity of simulated CH$_4$ to emissions and sinks, for comparison with $y$. $\mathbf{B}$ and $\mathbf{R}$ represent the prior and observation error covariance matrix, respectively. The cost function $J$ is minimized iteratively by the M1QN3 algorithm (Gilbert and Lemaréchal, 1989). We do not include sinks in the control vector $x$ but prescribe the different OH fields mentioned above.

Prior knowledge ($x^b$) on CH$_4$ emissions is provided by the Global Carbon Project (GCP, Saunois et al., 2019). The GCP emission inventory includes time-varying anthropogenic and fire emissions and climatology of the natural emissions. Global total CH$_4$ emissions of the GCP inventory are 511 Tg yr$^{-1}$ in 2000, increased to 562 Tg yr$^{-1}$ in 2010, and 581 Tg yr$^{-1}$ in 2016 (with soil uptake excluded). The soil uptake of CH$_4$ is estimated to be 38 Tg yr$^{-1}$ with seasonal variations. Averaged over 2000-2016, the anthropogenic sources (including biofuel emissions, agriculture, and waste) and wetlands contribute 56% and 32% of total CH$_4$ emissions, respectively (Fig. S1). The prior information of emissions by sector in each grid cell is used to separate the total optimized CH$_4$ emissions into four broad categories: wetlands, agriculture and waste, fossil fuel, and other natural sources (biomass burning, termite, geological, and ocean emissions). The spatial distributions of the prior emissions from the four categories averaged over 2000-2016 are shown in Fig. S1. A detailed description of the GCP emission inventory can be found in Zhao et al. (2019) and Saunois et al. (2019). The prior error of CH$_4$ fluxes is set to 100% of $x^b$, and the error correlation is calculated with a correlation length of 500 km over land and 1000 km over the oceans for CH$_4$ fluxes.

The vector of observations ($y$) is generated from surface measurements of the World Data Centre for Greenhouse Gases (WDCGG, https://gaw.kishou.go.jp/) through the WMO Global Atmospheric Watch (WMO-GAW) program. The surface measurements include both continuous time series of hourly afternoon observations and flask data. In total, 97 sites are included here, covering different time periods,
including 68 sites from the Earth System Research Laboratory from the U.S. National Oceanic and Atmospheric Administration (NOAA/ESRL, Dlugokencky et al. (1994)), 14 sites from the Laboratoire des Sciences du Climat et de l'Environnement (LSCE), 8 sites from Environment and Climate Change Canada (ECCC), 4 sites from the Commonwealth Scientific and Industrial Research Organisation (CSIRO, Francey et al. (1999)), and 3 from the Japan Meteorological Agency (JMA: http://www.jma.go.jp/jma/indexe.html). The location of the sites is shown in Fig. S2.

Atmospheric CH₄ sensitivities to fluxes \( (H(x)) \) are simulated by LMDz5B, an offline version of the LMDz atmospheric model (Locatelli et al., 2015), which has been widely used for CH₄ studies (e.g. Bousquet et al., 2005; Pison et al., 2009; Lin et al., 2018; Zhao et al., 2019). LMDz5B is associated with the simplified chemistry module SACS (Pison et al., 2009), which calculates the CH₄ chemical sink using prescribed 4D OH and O\(^{(1D)}\) fields. Air mass fluxes simulated by the general circulation model LMDz with temperature and wind nudged to ERA Interim re-analysis meteorology data (Dee et al, 2011) are used to force the transport of chemical tracers in LMDz5B every 3 hours.

2.3 Model experiments

As shown in Fig. 1, we performed six groups of inversions (Inv1 to Inv6, 34 inversions in total). The impacts of OH on the top-down estimation of CH₄ emissions are comprehensively analyzed by comparing the inversion results within one group or between two different groups. We analyze the overall impacts of differences in OH burden, spatial distribution, and temporal change on CH₄ emissions (colored boxes on the right in Fig. 1), and separate the impacts of OH spatial distribution and temporal variations (colored boxes on the left in Fig. 1). The results are presented and discussed in three sections as shown in different colors in Fig. 1.

We perform four groups of 3-year CH₄ inversion experiments using 6 to 10 OH fields (Inv1 to Inv4, Fig.
1), and two groups of 17-year CH$_4$ inversions from 2000 to 2016 (Inv5 and Inv6, Fig. 1) using only CESM1-WACC M OH fields. For the short-term inversions, the first and last six months are treated as spin-up and spin-down periods and discarded from the following analyses (to avoid edge effect). Thus we only analyze the results over 2000/07-2002/06 (i.e. the early 2000s) for Inv1 and Inv2 and over 2007/07-2009/06 (i.e. the late 2000s) for Inv3 and Inv4. The early 2000s and the late 2000s represent the time periods with stagnant and resumed growth of atmospheric CH$_4$ mixing ratios, respectively. For the long-term inversions, we take a one-year spin-up and spin-down and analyze the 15-year results from 2001 to 2015.

The aim of Inv1, conducted for 2000-2002 with 10 OH fields, is to quantify the influence of both OH burden and spatial distributions on top-down estimates of global, regional and sectoral CH$_4$ emissions (the brown box with the solid line, Fig. 1). To separate the influence of OH spatial distributions from that of global mean [OH], we conduct Inv2, where all the prescribed OH fields are globally scaled to the global [OH]$_{GM}$-CH$_4$ value of the INCA NMHC OH field in 2000 to get the same loss of CH$_4$ by OH (scaled OH fields). By doing so, differences in inversions of Inv2 originate solely from OH spatial distributions (the brown box with the dashed line, Fig. 1).

To quantify the influence of OH on temporal CH$_4$ emission changes, we also conduct Inv3 and Inv4 over 2007-2009, with 6 scaled OH fields (instead of 10 to limit computational time). While both of the inversions are done for 2007-2009, the OH variations during 2007-2009 and 2000-2002 are used for the two inversions, respectively. Therefore, while differences of Inv3−Inv2 reveal the impact of OH on temporal CH$_4$ emission changes (E_change_all, the yellow box with solid lines of Fig. 1), Inv4−Inv2 and Inv3−Inv4 allow separating the impacts only from OH spatial distributions (E_change_fixoh) and temporal variations (E_change_varoh), respectively (the yellow boxes with dashed lines in Fig. 1).
Finally, we test the impact of OH year-to-year variations and trends on CH\textsubscript{4} emissions over the time period 2001-2015 by running two long-term inversions (Inv5 and Inv6) with the OH fields simulated by CESM-WACCM only (the green box with dashed lines in Fig. 1). Inv5 is forced by the OH fields with both year-to-year variations and trends, while Inv6 is forced by the OH fields for the year 2000. For each group, only one experiment has done for computational reasons. We chose OH fields simulated by CESM1-WACCM because it shows the largest year-to-year OH variations during 2000-2010 among the CCMI OH fields (Zhao et al., 2019). Therefore, inversions using CESM1-WACCM OH are expected to yield an upper limit of the influence of OH variations on CH\textsubscript{4} emissions as seen from atmospheric chemistry models.

In the following sections, to quantify uncertainties in top-down estimations of CH\textsubscript{4} emissions due to OH, we calculate OH-induced CH\textsubscript{4} emission differences as the standard deviation of inversion results with different OH fields.

3 Results

3.1 The impacts of OH burden and spatial distributions on CH\textsubscript{4} emissions in 2000-2002

3.1.1 Global total CH\textsubscript{4} emissions

Based on the ensemble of the 10 different OH fields listed in Table 1, global total emissions inverted by our system in Inv1 vary from 518 to 757Tg CH\textsubscript{4} yr\textsuperscript{-1} during the early 2000s (2000/07-2002/06), nearly covering the CH\textsubscript{4} emission range derived from previous bottom-up and top-down GCP syntheses (Kirschke et al., 2013; Saunois et al., 2016) (Table 2 and Fig. 2). The highest CH\textsubscript{4} emissions exceed 700Tg yr\textsuperscript{-1} are calculated using MOCAGE and SOCOL3 OH fields, for which [OH]\textsubscript{GM-CH\textsubscript{4}} (15.0×10\textsuperscript{5} and 16.3×10\textsuperscript{5} molec cm\textsuperscript{-3}) are much higher than those of other OH fields (10.3-12.6×10\textsuperscript{5} molec cm\textsuperscript{-3}), leading to a larger CH\textsubscript{4} sink, and as a consequence also larger CH\textsubscript{4} emissions due to the mass balance constraint of atmospheric inversions. The discrepancies between top-down and bottom-up estimated CH\textsubscript{4} emissions
are mainly attributable to the overestimation of natural sources (Kirschke et al., 2013) because the OH fields used in those top-down inversions were usually constrained by MCF, and most of them used the TransCom OH field. However, the [OH]_{GM-M} constrained by MCF still have uncertainties of more than ±10% (Prinn et al., 2001; Bousquet et al., 2005). If we exclude the MOCAGE and SOCOL3 OH fields, the multi-model estimated [OH]_{GM-M} is 10.9 ±0.9 \times 10^5 \text{molec cm}^{-3} (\text{with uncertainty less than} ±10\%), which leads to global total CH$_4$ emissions ranging from 518 to 611 Tg yr$^{-1}$ in Inv1. This range is about twice the range given by previous top-down inversions (Kirschke et al., 2013; Saunois et al, 2016). Our results indicate that considering different OH fields generated by different models within top-down CH$_4$ inversions would help decreasing discrepancies with bottom-up estimations while leading to larger uncertainty on the CH$_4$ budget.

Plotting top-down estimated CH$_4$ emissions against [OH]_{GM-CH4}, which directly reflects the global OH oxidizing efficiency with respect to CH$_4$ (Lawrence et al., 2001), reveals that the global total CH$_4$ emissions vary linearly with [OH]_{GM-CH4} (r$^2$ >0.99, Fig. 2, right panel). The top-down estimation of global total CH$_4$ emissions (EMIS$_{CH4}$) can be approximately calculated as:

$$\text{EMIS}_{CH4}=40.4\times[OH]_{GM-CH4}+66.7$$

(1)

Where [OH]_{GM-CH4} is given in unit $10^5$ molec cm$^{-3}$ and EMIS$_{CH4}$ in Tg yr$^{-1}$. Using box-model inversions, previous studies calculated that a 4% ($0.4\times10^5$molec cm$^{-3}$) decrease in [OH]$_{GM}$ is equivalent to an increase of 22 Tg yr$^{-1}$ CH$_4$ emissions (Rigby et al., 2017; Turner et al., 2017, 2019). If we apply the same [OH]$_{GM}$ changes in Eq.1 ($0.4\times10^5$molec cm$^{-3}$), the equivalent emissions change is 16 Tg yr$^{-1}$, about 25% smaller than that given by Turner et al., (2017). This difference probably results from the different treatment of inter-hemispheric transport and stratospheric CH$_4$ loss in global 3D transport models compared to the simplified box-models (Naus et al., 2019).

### 3.1.2 Regional CH$_4$ emissions
Because of the long lifetime of CH$_4$ relative to OH, the top-down estimates of regional CH$_4$ emissions can be influenced by both global total OH burden and OH spatial distributions. In Inv2 we remove the influence of the global total OH burden by scaling all the OH fields to get the same CH$_4$ loss as calculated by INCA NMHC in 2000. As such, Inv2 provides the uncertainty range of CH$_4$ emissions induced by OH spatial distribution in both horizontal and vertical directions when assuming that the global total burden of OH can be precisely constrained. Thus, Inv1 (the inversions using original OH fields) and Inv2 (the inversions using scaled OH fields) yield upper (uncertainties from both global OH burden and spatial distributions) and lower (uncertainties only from OH spatial distributions) limits of influences of OH on regional CH$_4$ emissions, respectively. Since MOCAGE and SOCOL3 OH fields show much higher [OH]$_{GM}$ than constrained by MCF observations (~10$\times$10$^5$ molec cm$^{-3}$, e.g. Prinn et al., 2001; Bousquet et al., 2005) and give much higher estimates of CH$_4$ emissions (>700 Tg yr$^{-1}$) than other OH fields, we exclude inversion results with these two OH fields from the following analyses.

In response to both global total OH burden and inter-hemispheric OH ratios (Table 1), CH$_4$ emissions over northern and southern hemispheres calculated by Inv1 (Table 2) vary from 368 to 424 Tg yr$^{-1}$ (401 ± 21 Tg yr$^{-1}$) and 138 to 187 Tg yr$^{-1}$ (166 ± 15 Tg yr$^{-1}$), respectively, resulting in a range in inter-hemispheric CH$_4$ emission difference (NH − SH) of 206-254 Tg yr$^{-1}$ (236 ± 14 Tg yr$^{-1}$). When scaling all OH fields to the same loss for Inv2, the standard deviations of hemispheric CH$_4$ emissions are reduced to 7 Tg CH$_4$ yr$^{-1}$ for both hemispheres, much smaller than those derived in Inv1 (21 Tg yr$^{-1}$ and 15 Tg yr$^{-1}$ over the northern and southern hemisphere, respectively). However, the CH$_4$ emission inter-hemispheric difference calculated by Inv2 remains at 236 ± 14 Tg yr$^{-1}$, similar to that calculated by Inv1, which indicates that the hemispheric CH$_4$ emissions differences are mainly determined by OH spatial distributions. Without the TransCom OH simulation, the inter-hemispheric CH$_4$ emission difference ranges between 232 and 246 Tg yr$^{-1}$. The TransCom OH field, for which OH N/S ratio is 1.0, leads to an inter-hemispheric CH$_4$ emission difference of 205 Tg yr$^{-1}$, 35 Tg yr$^{-1}$ (27 Tg yr$^{-1}$) smaller than the mean (minimum) inter-hemispheric
difference calculated using other OH fields (OH N/S ratio = 1.2-1.3). Previous studies show that differences in atmospheric transport models can lead to ±28Tg yr\(^{-1}\) uncertainties in the top-down calculation of the inter-hemispheric CH\(_4\) emission difference, using a single OH field – TransCom (Locatelli et al., 2013). Here, using a single atmospheric transport model, but different OH fields, we find a ±14Tg yr\(^{-1}\) uncertainty, about half of the atmospheric transport model uncertainty. Combining the two studies, we could expect more than 30Tg yr\(^{-1}\) uncertainty in top-down estimates of the inter-hemispheric CH\(_4\) emission difference, based on different atmospheric models and knowledge on OH.

Fig. 3 shows the optimized and prior CH\(_4\) emissions calculated by Inv1 (top) and Inv2 (bottom) over latitudinal intervals (left panels) and large emitting regions (right panels). Compared with prior emissions, nearly all the optimized latitudinal and regional emissions show the same increment direction from prior emissions, but the magnitudes of the increment largely vary. The CH\(_4\) emissions calculated by Inv1 amount to i) 147±14Tg yr\(^{-1}\) and are 1-47Tg yr\(^{-1}\) higher than the prior over the southern tropical regions (30°S-0°), ii) 199±14Tg yr\(^{-1}\) and are 6-45Tg yr\(^{-1}\) higher than the prior over the northern tropical regions (0°-30°N), and iii) 174±8Tg yr\(^{-1}\) and are 1-26Tg yr\(^{-1}\) lower than the prior over the northern mid-latitude regions (30°N-60°N). The regional emission standard deviations due to OH correspond to 11\%, 8\%, and 4\% of prior emissions over the southern tropical, northern tropical, and northern hemisphere mid-latitude regions, respectively. This range of uncertainty is comparable to those from different inversions given by Saunois et al. (2016). Over the large emitting regions Europe (EU), Canada (CAN), and China (CHN), optimized emissions are lower than the prior. The Inv1 calculated emissions show the largest absolute OH induced differences over northern South America (NSA, 73±9Tg yr\(^{-1}\)), South Asia (SAS, 59±6Tg yr\(^{-1}\)), and China (42±5Tg yr\(^{-1}\)) (Fig. 3, right panels and Table 3), which account for 15-10\% of the prior emission over the corresponding regions. At the model grid-scale, the uncertainties due to OH can be much larger than the regional mean (middle panel of Fig. 3), for example, larger than 20\% of the prior emissions over South America and central Africa, and larger than 15\% of the prior over East and South
We now compare the inversion results using the original OH fields (Inv1) with those using scaled OH fields (Inv2) to estimate how much the optimized regional CH$_4$ emission differences of Inv1 are dominated by OH spatial distributions versus the global OH burden. Applying one single global scaling factor per model reduces the inter-model differences of original OH fields by 33%, 67% and 33% in the southern tropics, northern tropics, and northern mid-latitudes (30°-90°N) (Table S1). This scaling results in 57%, 93%, and 22% reduction of OH induced latitudinal CH$_4$ emission differences, respectively (Fig. 3, left panels, comparing standard deviations of Inv1 and Inv2). At the regional scale (Fig. 3, right panel and Table 3), the OH spatial distribution-induced CH$_4$ emission differences (standard deviation of Inv2) account for 50% of total differences over northern mid-latitude regions (China, North America) and southern tropics (South America, with largest inter-model differences of scaled OH, Fig. S3). Over northern tropical regions (Southern Asia and Southeast Asia), the OH spatial distribution induces negligible CH$_4$ emission differences.

The comparison of Inv1 and Inv2 reveals that tropical regions are less sensitive to OH spatial distribution than mid and high latitude regions in our framework. One possible explanation could be the location of monitoring sites. Over tropical regions, surface monitoring sites are mainly located in remote places, but for 30°N-90°N, there are several monitoring sites located at or near the regions with high CH$_4$ emission rates and OH uncertainties (e.g. North America, Europe, and downwind of East Asia). Because they are more constrained by monitoring sites near source regions, emissions of 30°-90°N are found to be more sensitive to the OH spatial distributions whereas emissions over tropical regions are only constrained by remote sites and are found to be more sensitive to the global OH burden.
3.1.3 Global and regional CH$_4$ emissions per source category

Fig. 4 compares optimized and prior global total CH$_4$ emissions and the difference between the prior and optimized CH$_4$ emissions for four broad source categories: wetlands, agriculture and waste (Agri-waste), fossil fuels, and other. We attribute the optimized emissions to different source sectors depend on the relative strength of individual prior sources in each grid-cell. With original OH fields, Inv1 calculates CH$_4$ emissions of 203±15Tg yr$^{-1}$ for wetlands, 209±12Tg yr$^{-1}$ for Agri-waste, 89±4Tg yr$^{-1}$ for fossil fuel, and 66±3Tg yr$^{-1}$ for other natural sources. Optimized emissions of the four sectors are 23±15Tgyr$^{-1}$(-2-42Tg yr$^{-1}$), 13±12Tg yr$^{-1}$(-3-29Tg yr$^{-1}$), 5±4Tg yr$^{-1}$(-1-9Tg yr$^{-1}$), and 4±3Tg yr$^{-1}$(-0.1-8Tg yr$^{-1}$) higher than the prior emissions, respectively. Although Inv2 is conducted with scaled OH fields and all inversions calculate similar global total CH$_4$ emissions (551±2Tg yr$^{-1}$), optimized CH$_4$ emissions still show some uncertainties due to OH (as standard deviation) (3Tg yr$^{-1}$ for wetland emissions and 2Tg yr$^{-1}$ for agriculture and waste, yellow boxplots in Fig. 4), in response to different OH spatial distributions.

We have further calculated CH$_4$ emissions per source category and per region estimated by Inv1 (Fig. 5), to explore the contribution of each region to the OH-induced sectoral emission uncertainties. Wetland CH$_4$ emissions mainly dominate emissions over Northern South America, Africa, South and East Asia, and Canada. Northern South America (53±7Tg yr$^{-1}$) and Africa (30±2Tg yr$^{-1}$) contribute most of the global total wetland OH induced emission differences and are 1-22Tg yr$^{-1}$ and 1-8Tg yr$^{-1}$ higher than prior emissions, respectively. In contrast to the higher wetland emissions than prior ones over tropical regions, optimized boreal wetland emissions (Canada) are 6-9Tg yr$^{-1}$ lower than prior emissions, consistent with lower top-down estimations than the prior given by Saunois et al. (2016). Agriculture and waste emissions are most intensive over China (25±3Tg yr$^{-1}$) and South Asia (SAS) (39±3Tg yr$^{-1}$). The optimized inventories show lower agriculture and waste emissions over China (0.6-10Tg yr$^{-1}$) and Europe (1-3Tg yr$^{-1}$) and much higher emissions over SAS (4-13Tg yr$^{-1}$) compared with the prior emission inventory. The OH induced differences in fossil fuel emissions are found mainly in China and Africa, which are 0.8-5Tg
yr\(^{-1}\) lower and 0.6-3Tg higher than prior emissions, respectively. In agreement with the previous regional discussion, scaling the OH (Inv2) highly reduces the uncertainties attributable to different OH over the tropical regions but not for the mid-high latitude regions. In Inv2, the largest CH\(_4\) emission differences due to different OH spatial distribution are found for wetland emissions in South America (60±4Tg yr\(^{-1}\)), agriculture and waste emissions in South Asia (17±1Tg yr\(^{-1}\)) and China (24±2Tg yr\(^{-1}\)), and fossil fuel emissions in China (8±0.7Tg yr\(^{-1}\)) and Russia (9±0.4Tg yr\(^{-1}\)).

Previous studies have highlighted that anthropogenic emissions over China are largely overestimated by bottom-up emissions inventories compared with top-down estimates (Kirschke et al., 2013; Tohjima et al., 2014; Saunois et al., 2016). In our study, total anthropogenic emissions (agriculture, waste, and fossil fuel) over China are 1-15Tg yr\(^{-1}\) lower than the prior bottom-up inventory as calculated by Inv1, and 7-14Tg yr\(^{-1}\) as calculated by Inv2, with the lowest emissions calculated with the TransCom OH field (for both Inv1 and Inv2). The TransCom OH field is the one most widely used in current top-down CH\(_4\) emission estimations but shows much lower [OH] over China than other OH fields (Zhao et al., 2019), which may be due to the use of the same NO\(_x\) profile over East Asia as for remote regions based on the observations of 1990s when constructing the TransCom OH field (Spivakovsky et al., 2000). Thus, the large reduction of top-down estimated anthropogenic CH\(_4\) emissions over China as compared to the prior emissions may be partly due to an underestimation of [OH] over China in the TransCom field.

### 3.2 Impact of OH on CH\(_4\) emission changes between 2000-2002 and 2007-2009

As shown in Table 4, the global mean [OH] simulated by CCMI models increased by 0.7%-1.8% from 2000-2002 to 2007-2009, in response to anthropogenic emissions and climate change (Zhao et al., 2019), whereas the INCA-NMHC model-simulated global [OH] shows a slight decrease of 0.5%. The TransCom OH field, being constant over time, shows no change. The global scale increase mainly results from the combination of a higher increase in the tropics compared to the northern extra-tropics and a slight decrease
in the southern extra-tropics. As a result, the changes in OH between the two periods show different patterns between regions. We have conducted inversions for 2007-2009 with scaled OH fields (Inv3) to explore how uncertainties in OH (both spatial distribution and temporal changes) can influence the top-down estimation of temporal CH₄ emission changes from the early 2000s (2000/07-2002/06) to the late 2000s (2007/07-2009/06) (E_change_all, Inv3—Inv2). We have also performed Inv4 for 2007-2009 but using OH fields of 2000-2002 to separate the differences due to different time periods, with obviously different emissions (E_change_fixoh, Inv4 — Inv2) to the ones due to temporal changes in OH (E_change_varoh, Inv3—Inv4).

### 3.2.1 Global total emission changes between 2000-2002 and 2007-2009

**Total emission changes.** Changing the inversion years and OH fields increase CH₄ emissions by 21.9±5.7Tg yr⁻¹ (E_change_all, 16.6-30.0Tg yr⁻¹, Table 5) from the early 2000s (Inv2) to the late 2000s (Inv3). The largest CH₄ increase of 30.0Tg yr⁻¹ is estimated with CESM1-WACCM OH fields (for which OH increased by 1.8% from 2000-2002 to 2007-2009), 13.4Tg yr⁻¹ higher than the smallest increase of 16.6 yr⁻¹ from the INCA NMHC OH field (for which OH decreased by 0.5% from 2000-2002 to 2007-2009). In Saunois et al. 2017, the min-max uncertainty range of emission changes between 2002-2006 and 2008-2012 was 16Tg yr⁻¹. This means that the uncertainty attributable to different OH spatial distribution and temporal changes(13.4Tg yr⁻¹), is comparable to the min-max uncertainty resulting from using different atmospheric chemistry transport models and observations (surface and satellite), but mostly constant OH over time(16Tg yr⁻¹, Saunois et al., 2017).

**Spatial versus temporal OH effects.** Keeping OH fields from 2000-2002 (Inv4 — Inv2), top-down estimated CH₄ emissions increase only by 16.9±1.9Tg yr⁻¹ between the early 2000s to the late 2000s (E_change_fixoh, 14.3-19.3Tg yr⁻¹, Table 5). Only changing OH from 2000-2002 to 2007-2009 (Inv3—Inv4), top-down estimated CH₄ emissions over the late 2000s change by +5.1±6.4Tg yr⁻¹
(E_change_varoh, -2.7-13.5Tg yr⁻¹, Table 5) due to OH temporal variations. By comparing E_change_all, E_change_fixoh, and E_change_varoh, we estimate that, on average, OH temporal variations (E_change_varoh, 5.1±6.4Tg yr⁻¹) contribute 25% of total optimized emission changes between the early and late 2000s (E_change_all, 21.9±5.7Tg yr⁻¹, Table 5). As listed in Table 5, the largest emission increase due to OH temporal variations is calculated using MRI-ESM1r1 OH fields, for which a 1.1% global increase in OH can up to double the top-down estimation of CH₄ increased from the early to the late 2000s. This result indicates that a large bias likely exists in the former top-down estimation of the CH₄ emission trend calculated without considering OH changes (Saunois et al., 2017).

Comparing E_change_all, E_change_fixoh, and E_change_varoh also allows separation of uncertainties in OH spatial distribution from uncertainties in OH temporal variations. Overall, the uncertainties in OH spatial distributions and OH temporal changes lead to 1.9Tg yr⁻¹ (standard deviation of E_change_fixoh) and 6.4Tg yr⁻¹ (standard deviation of E_change_varoh) uncertainties in optimized CH₄ emission changes from the early 2000s to late 2000s (Table 5), respectively. These results indicate that the OH spatial distribution, which is not considered in box-models, can also contribute to the uncertainties in optimized CH₄ emission changes.

3.2.2 Emission changes by source types and regions

Total emission changes. We further analyze the influence of OH (both spatial distributions and temporal variations) on the top-down estimated sectoral and regional CH₄ emission changes from the early 2000s to the late 2000s (E_change_all, Inv3—Inv2). As shown in Fig. 6 (top panels), the smaller increase of the optimized global CH₄ emissions from the early 2000s to the late 2000s (E_change_all, 21.9±5.7Tg yr⁻¹) compared to the prior change (39.4Tg yr⁻¹) is mainly due to decrease in wetland emissions over the southern tropics (-4.4±1.5Tg yr⁻¹, 15°S-0°) and northern mid-latitude regions (-3.4±0.4Tg yr⁻¹, 45°N-60°N) in contrast to climatology prior wetland emissions, and a lower fossil fuel emission increase over
30°N-45°N (3.7 ± 0.4 Tg yr\(^{-1}\)) compared to prior emission increase (8.9 Tg yr\(^{-1}\)). Wetlands (-3.5 ± 2.5 Tg yr\(^{-1}\)) and agriculture and waste (14.2 ± 2.1 Tg yr\(^{-1}\)) contribute most of the total OH induced uncertainty in global total emission changes (21.9 ± 5.7 Tg yr\(^{-1}\)) from the early 2000s to the late 2000s (Table 6), whereas fossil fuel emissions (8.7 ± 0.8 Tg yr\(^{-1}\)) show smaller uncertainty.

Considering emissions over latitudinal bands (Fig. 6, top panels), the largest spread of emission changes are found over the southern tropics (15°S–tropics, -1.3 to -6.5 Tg yr\(^{-1}\)), northern subtropics (15°N-30°N, 15.5 to 20.0 Tg yr\(^{-1}\)), and northern extratropical regions (30°N-45°N, 7.6 to 10.7 Tg yr\(^{-1}\)). The spread over the southern tropics is dominated by emission changes from wetlands (-2.3 to -6.3 Tg yr\(^{-1}\)), over northern subtropics by agriculture and waste (7.3 to 10.0 Tg yr\(^{-1}\)), and over northern extratropical regions by agriculture and waste (3.7 to 5.1 Tg yr\(^{-1}\)) and fossil fuels (3.3 to 4.3 Tg yr\(^{-1}\)). At the regional scale (Fig. 7, top panels), northern South America (-1.2 to -5.2 Tg yr\(^{-1}\)), South Asia (9.1 to 12.4 Tg yr\(^{-1}\)), and China (-0.1 to 4.9 Tg yr\(^{-1}\)) show the largest differences in emission changes from the early 2000s to the late 2000s. The multi-inversions calculated emission changes in China disagree in sign (Fig. 7; top panels), mainly due to differences in the agriculture and waste sector, which range from 1.3 Tg yr\(^{-1}\) decrease to 1.5 Tg yr\(^{-1}\) increase from the early 2000s to the late 2000s. Previous studies have shown that large differences exist in different top-down estimated regional and sectoral emission changes mainly due to model transport errors (Saunois et al., 2017). Here, our results show that uncertainties due to OH spatio-temporal variations can also contribute to these biases in top-down inversions.

**Spatial versus temporal effects.** We compare regional and sectoral E\(_{\text{change\_all}}\) (Inv3−Inv2) with E\(_{\text{change\_fixoh}}\) (Inv4−Inv2) and E\(_{\text{change\_varoh}}\) (Inv3−Inv4) to separate influences of OH spatial distribution and temporal variations. As shown in the bottom panels of Fig. 6 and Fig. 7, at the regional scale, OH temporal variations mainly perturb top-down estimated CH\(_4\) emission changes (Fig. 6; bottom panel) over the southern tropics (0°-15°S, -3.4-5.9 Tg yr\(^{-1}\)) and northern subtropics (15°N-30°N, 0-5.4 Tg yr\(^{-1}\))
yr⁻¹). This corresponds to the two largest spreads observed in Fig. 7 (bottom panel) associated with wetland emissions over northern South America (-2.0-3.5Tg yr⁻¹) and with agriculture and waste emissions over South Asia (-0.2-2.6Tg yr⁻¹). Among the four emission sectors, wetland emissions (mainly southern tropical wetland) show the largest increase (2.1±3.4Tg yr⁻¹) in response to OH temporal changes (Table 6), which account for 60% of total wetland emission changes between these two periods.

The OH spatial distribution can lead to large uncertainties in regional CH₄ emission changes. For regional and latitudinal scales, the spreads (uncertainties) of E_change_fixoh (results from OH spatial distributions) (Fig. S4 and Fig. S5) are comparable to E_change_all (top panels of Fig. 6 and Fig. 7) as mentioned above (e.g., 2.6-5.4Tg yr⁻¹ decrease over northern South America, 6.1-11.1Tg yr⁻¹ increase over South Asia, and -0.4-4.2Tg yr⁻¹ increase over China). These results show that even if the global total OH burden is well constrained (as in Inv4 and Inv2, where all OH fields are scaled to the same [OH]GM-CH₄, and the differences in optimized CH₄ emissions changes from the early 2000s to the late 2000s are only due to different OH spatial distributions), top-down estimates of sectoral and regional temporal CH₄ emission changes remain highly uncertain.

3.3 Impacts of OH on year to year variations of CH₄ emissions from 2001 to 2015

To infer the influence of OH year-to-year variations on top-down optimized long-term CH₄ emission changes, we conducted two inversions, Inv5 and Inv6. Inv5 calculates optimized CH₄ emissions from 2001-2015 with the CESM1-WACCM OH field varying from one year to the next, while Inv6 uses the CESM1-WACCM OH field but fixed to the year 2000. The choice of the CESM1-WACCM OH field is explained in Sect. 2.3 above. As shown in Fig. 8, the [OH]GM-CH₄ of the CESM1-WACCM OH field increases by $0.47 \times 10^5$ molec cm⁻³ (4.2%) from 2001 to 2011 and then decreases by $0.13 \times 10^5$ molec cm⁻³ (1.1%) from 2011 to 2015.
With OH fixed to the year 2000 (Inv6), global CH$_4$ emissions stall at 550±2Tg yr$^{-1}$ during 2001-2003, decrease to 538Tg yr$^{-1}$ in 2004, which is different from the continuous increase of CH$_4$ emissions given by the bottom-up inventory (Fig. 8, top panel). After 2004, global total CH$_4$ emissions show a positive trend of 3.5 ± 1.8Tg yr$^{-2}$ (P<0.05), but smaller than the prior bottom-up inventory (4.3 ± 0.6Tg yr$^{-2}$ (P<0.05)). Both stalled/decreased emissions during 2001-2004 and increasing trend after 2004 are consistent with previous top-down estimations (Saunois et al., 2017).

The trend of global CH$_4$ emission during 2004-2016 calculated by Inv5 (using varying OH) is 4.8 ± 1.8Tg yr$^{-2}$ (P<0.05), which is 1.3Tg yr$^{-2}$ (36%) higher than that calculated by Inv6 (OH fixed to 2000) due to the small increase in [OH], and also 0.5Tg yr$^{-2}$ higher than the prior emission trend (4.3 ± 0.6Tg yr$^{-2}$). Accounting for the OH increasing trend leads to increasing the prior trend in Inv5 instead of decreasing it in Inv6. When calculating the differences between Inv5 and Inv6 for different latitude intervals, we find that before 2004, differences between Inv5 and Inv6 are mainly contributed by northern middle-latitude regions, whereas after 2004 they are dominated by tropical regions (Fig. 8, bottom).

We further compare CH$_4$ emission trends for the four previously defined emission sectors and the ten continental regions between Inv5 and Inv6. As shown in Fig. 9, the positive global CH$_4$ emission trend during 2004-2016 is mainly contributed by anthropogenic sources from agriculture and waste, and fossil fuel-related activities, which are 1.9±0.7Tg yr$^{-2}$, and 2.3±0.4Tg yr$^{-2}$, respectively, as calculated by Inv6 (fixed OH). Wetland emissions show a small negative trend (-0.5 ± 0.7Tg yr$^{-2}$) and other natural emissions do not show a significant trend (0.04±0.6Tg yr$^{-2}$). Both sectors show large uncertainties in their trends reflecting large year-to-year variations. When considering [OH] variations, Inv5 estimates a higher agriculture and waste emission trend (2.4 ± 0.8Tg yr$^{-2}$) compared to Inv6, mainly contributed by China (1.5±0.5Tg yr$^{-2}$ for Inv6 versus 1.7±0.5Tg yr$^{-2}$ for Inv5) and southern South America(-0.1±0.1Tg yr$^{-2}$ for Inv6 versus 0.1±0.3Tg yr$^{-2}$ for Inv5). Accounting for temporal OH variations the negative wetland
emission trend reduces to near zero (0.1±0.6 Tg yr⁻²), mainly due to increased emission trends over northern South America (-0.3±0.3 Tg yr⁻² for Inv6 versus 0.2±0.5 Tg yr⁻² for Inv5). In contrast to agriculture and waste, and wetland emissions, fossil fuel emissions have a similar positive trend of 2.4±0.4 Tg yr⁻² in Inv5 and Inv6. This results from a higher CH₄ emission trend over China calculated by Inv5 balanced by a lower CH₄ emission trend over North America and Russia (0.2 ± 0.2 Tg yr⁻² for Inv6 versus 0.1 ±0.3 Tg yr⁻² for Inv5) since the CESM1-WACCM OH field shows a significant negative [OH] trend over North America (Zhao et al., 2019).

4 Conclusions and discussion

In this study, we have performed six groups of variational Bayesian inversions (top-down, 34 inversions in total) using 10 different prescribed OH fields to quantify the influence of OH burden, temporal variations, and spatial distributions on the top-down estimation of i) global total, regional, and sectoral CH₄ emissions, ii) emission changes between the early 2000s and late 2000s, and iii) year-to-year emission variations. The top-down system estimates CH₄ emissions by assimilating surface observations with atmospheric transport of CH₄ calculated by the offline version LMDz5B of the LMDz atmospheric model using different prescribed OH fields.

Based on the ensemble of 10 original OH fields ([OH]GM-CH₄: 10.3-16.3×10⁵ molec cm⁻³), the global total CH₄ emissions inverted by our system in Inv1 vary from 518 to 757 Tg yr⁻¹ during the early 2000s, nearly covering the CH₄ emission range calculated by previous bottom-up and top-down GCP syntheses (Kirschke et al., 2013; Saunois et al, 2016). The top-down estimated global total CH₄ emission varies linearly with [OH]GM-CH₄, and we found that 1×10⁵ molec cm⁻³ (10%) higher [OH]GM-CH₄ can result in 40.4 Tg yr⁻¹ changes in optimized CH₄ emissions, which is smaller than changes inferred from box-models most probably due to inherent simplifications in the representation of atmospheric transport and chemistry.
in these models.

At regional scale (excluding the two highest OH fields), OH-induced CH$_4$ emission uncertainties are largest over northern South America (12%, standard deviation/mean), South Asia (10%), and China (12%), leading to significant uncertainties in optimized emissions from the wetland and agriculture and waste sectors. By performing inversions with a globally-scaled OH field, we calculated that emission uncertainties due to different OH spatial distributions account for ~50% of total uncertainties (induced by both different OH burden and different OH spatial distributions). CH$_4$ emission differences due to OH spatial distributions are the largest in northern South America and China but are negligible over South Asia and other northern tropical regions. Our study shows that tropical regions appear less sensitive to OH spatial distribution than mid-to-high latitude regions based on CH$_4$ emission optimization with surface observations.

The global CH$_4$ emission change between 2000-2002 and 2007-2009 as estimated by top-down inversions using 6 different OH fields, is 21.9±5.7Tg yr$^{-1}$, of which 25% (5.1±6.4Tg yr$^{-1}$) is contributed by OH temporal variations (mainly by an increase in [OH]), while 75% can be attributed to emission changes resulting from increase in observed CH$_4$ mixing ratios (constant OH). Among the four emission sectors, wetland emissions (mainly southern tropical wetlands) show the largest increase of 2.1±3.4Tg yr$^{-1}$ in response to OH temporal changes, which account for 60% of total wetland emission changes between these two periods. For global total emission changes, OH spatial distributions lead to lower uncertainties than temporal variations (1.9Tg yr$^{-1}$ versus 6.4Tg yr$^{-1}$), but on the regional scale, OH spatial distributions and temporal variations are of equal importance for quantifying CH$_4$ emission changes.

As the modeled OH used here mainly shows an increase in [OH] (meaning increasing CH$_4$ sink) during the 2000s, our inversion using year-to-year OH variations infers a 36% higher CH$_4$ emission trend.
compared with an inversion driven by climatological OH over the 2001-2015 period. The different OH fields from CCMI models consistently show increasing OH trends during 2000-2010 (Zhao et al., 2019). These variations disagree with MCF-constrained [OH], which show a decrease after 2003/2004 (Rigby et al., 2017; Turner et al., 2017). A drop of OH between 2006-2007 (Rigby et al., 2008, Bousquet et al., 2011) is captured by CESM1-WACCM OH fields but with (possibly) smaller changes (1%) compared to (the very uncertain) 4±14% constraint by MCF (Rigby et al., 2008). This OH drop in 2006-2007 results in a 6Tg yr\(^{-1}\) smaller increase of CH\(_4\) emissions between 2006 and 2007 compared to that derived using constant OH. However, such [OH] drop is treated as a year-to-year variation instead of a trend, and cannot fully explain the resumption of CH\(_4\) growth from 2006-2007. Thus, during 2004-2010, at the decadal timescale, if the CCMI models represent the OH trend properly, a higher increasing trend of CH\(_4\) emissions is needed to match the CH\(_4\) observations (compared to the CH\(_4\) emission trend derived using constant OH). After 2010, CCMI models simulated different OH trend (Zhao et al., 2019), thus the influence on CH\(_4\) emission trend is more uncertain.

Compared to previous box-model studies (e.g. Rigby et al., 2017; Turner et al. 2017), the inversions performed in this study take advantage of 4D OH fields from CCMI to better quantify impacts on regional and sectoral emission estimations. The results indicate that OH spatial distributions, which are difficult to obtain from MCF observations, are equally important than the OH burden for constraining CH\(_4\) emissions over mid- and high- latitude regions. Also, Spatio-temporal OH variations are critical to properly infer regional and sectoral CH\(_4\) emission temporal changes.

Top-down inversions, particularly variational Bayesian systems, are powerful tools to infer greenhouse gas budgets, in particular, methane the target of this study. However, they suffer from some limitations impacting the budget uncertainty. Some work has been done regarding atmospheric transport errors (e.g., Locatelli et al., 2013, 2015) and sensitivity to observation constraints (Locatelli et al., 2015; Houweling
et al., 2000), but less on the chemistry side of the budget.

Overall, our study significantly contributes to assessing the impact of OH uncertainty on the CH\textsubscript{4} budget. We have shown that it is insufficient to consider a unique OH field, constant over time, in order to fully understand and assess the global CH\textsubscript{4} budget and its changes over time. Indeed, further work is needed to help determining OH fields to be used in future variational top-down inversion studies to properly account for changes in both source and sinks. There are different ways to optimize current OH fields. One way is conducting multi-species variational inversion of OH (e.g. Zheng et al., 2019) with such HFC species (Liang et al., 2017), formaldehyde (Glenn et al., 2019), CH\textsubscript{4} (Zhang et al., 2018; Maasakkers et al., 2019), or CO (Zheng et al., 2019); another way can be to build semi-empirical OH fields by combining atmospheric chemistry models and observation-based meteorological data and chemical species concentrations (e.g. NO\textsubscript{x}, CO, VOCs, etc) as initiated in Spivakovsky et al.(2000). These OH fields should include a mean 3D global [OH] distribution, associated with temporal variations and uncertainties. A lot remains to be done to better constrain the chemistry side of the global methane budget, a critical step toward its closure.

**Data availability**
The CCMI OH fields are available at the Centre for Environmental Data Analysis (CEDA; http://data.ceda.ac.uk/badc/wcrp-ccmi/data/CCMI-1/output; Hegglin and Lamarque, 2015), the Natural Environment Research Council's Data Repository for Atmospheric Science and Earth Observation. The CESM1-WACCM outputs for CCMI are available at http://www.earthsystemgrid.org (Climate Data Gateway at NCAR, 2019). The surface observations for CH\textsubscript{4} inversions are available at the World Data Centre for Greenhouse Gases (WDCGG, https://gaw.kishou.go.jp/, 2019). Other datasets, including INCA OH fields and optimized CH\textsubscript{4} emissions, can be accessed by contacting the corresponding author.

**Author contributions**
YZ, MS, and PB designed the inversion experiments, analyzed results, and wrote the manuscript. BZ
and XL helped with data preparation and inversion setup. JC and RJ discussed the results. AB developed the LMDz code for running CH₄ inversions. MH provided the CCMI OH fields. DW, ED, MR, and RL provided the atmospheric in situ data. All coauthors commented on the paper.

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### Tables

**Table 1.** Global tropospheric mean [OH] and inter-hemispheric OH ratios (N/S) averaged over 2000-2002 for 10 OH fields used in this study. The global tropospheric [OH] weighted by reaction with CH₄ ([OH]_{GM-CH₄}) and weighted by dry air-mass ([OH]_{GM-M}) are both given.

<table>
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<th>[OH]_{GM-M}</th>
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<tr>
<td>CESM1-WACCM</td>
<td>11.9</td>
<td>11.4</td>
<td>1.3</td>
</tr>
<tr>
<td>CMAM</td>
<td>12.2</td>
<td>11.3</td>
<td>1.2</td>
</tr>
<tr>
<td>EMAC-L90MA</td>
<td>11.8</td>
<td>11.5</td>
<td>1.2</td>
</tr>
<tr>
<td>GEOSCCM</td>
<td>12.6</td>
<td>12.3</td>
<td>1.2</td>
</tr>
<tr>
<td>MOCAGE</td>
<td>15.0</td>
<td>12.5</td>
<td>1.5</td>
</tr>
<tr>
<td>MRI-ESM1r1</td>
<td>10.9</td>
<td>10.6</td>
<td>1.2</td>
</tr>
<tr>
<td>SOCOL3</td>
<td>16.3</td>
<td>14.4</td>
<td>1.5</td>
</tr>
<tr>
<td><strong>Mean±SD</strong></td>
<td><strong>12.3±3.8</strong></td>
<td><strong>11.4±2.8</strong></td>
<td><strong>1.3±0.3</strong></td>
</tr>
<tr>
<td><strong>Mean±SD (8 OH)¹</strong></td>
<td><strong>11.3±0.8</strong></td>
<td><strong>10.9±0.9</strong></td>
<td><strong>1.2±0.1</strong></td>
</tr>
</tbody>
</table>

¹ The OH fields simulated by SOCOL3 and MOCAGE are excluded.

**Table 2.** The global total and hemispheric CH₄ emissions calculated with the 10 original OH fields (Inv1), and inter-hemispheric difference of CH₄ emissions calculated by Inv1 and Inv2 (scaled OH fields) during the early 2000s (2000/07/01-2002/06/01).

<table>
<thead>
<tr>
<th>Model</th>
<th>Global</th>
<th>0-90°N</th>
<th>90°S-0</th>
<th>N-S_{Inv1}</th>
<th>N-S_{Inv2}</th>
</tr>
</thead>
<tbody>
<tr>
<td>Prior</td>
<td>522</td>
<td>384</td>
<td>138</td>
<td>246</td>
<td>246</td>
</tr>
<tr>
<td>TransCom</td>
<td>530</td>
<td>368</td>
<td>162</td>
<td>206</td>
<td>205</td>
</tr>
<tr>
<td>INCA NMHC-AER-S</td>
<td>518</td>
<td>380</td>
<td>138</td>
<td>242</td>
<td>245</td>
</tr>
<tr>
<td>INCA NMHC</td>
<td>552</td>
<td>392</td>
<td>160</td>
<td>232</td>
<td>232</td>
</tr>
<tr>
<td>CESM1-WACCM</td>
<td>587</td>
<td>420</td>
<td>166</td>
<td>254</td>
<td>249</td>
</tr>
<tr>
<td>CMAM</td>
<td>599</td>
<td>419</td>
<td>180</td>
<td>239</td>
<td>245</td>
</tr>
<tr>
<td>EMAC-L90MA</td>
<td>589</td>
<td>414</td>
<td>175</td>
<td>239</td>
<td>237</td>
</tr>
</tbody>
</table>
Table 3. Regional CH$_4$ emissions (mean±SD) calculated by Inv1 and Inv2, and for the prior emissions during the early 2000s (2000/07/01-2002/06/01) in Tg yr$^{-1}$.

<table>
<thead>
<tr>
<th>Region</th>
<th>Inv1</th>
<th>Inv2</th>
<th>Prior</th>
</tr>
</thead>
<tbody>
<tr>
<td>North America(USA)</td>
<td>45±2</td>
<td>45±1</td>
<td>42</td>
</tr>
<tr>
<td>Canada(CAN)</td>
<td>27±1</td>
<td>27±1</td>
<td>33</td>
</tr>
<tr>
<td>Europe(EU)</td>
<td>27±1</td>
<td>27±1</td>
<td>29</td>
</tr>
<tr>
<td>Russia(RUS)</td>
<td>33±1</td>
<td>33±1</td>
<td>31</td>
</tr>
<tr>
<td>China(CHN)</td>
<td>42±5</td>
<td>40±3</td>
<td>51</td>
</tr>
<tr>
<td>Southeast Asia(SEAS)</td>
<td>38±3</td>
<td>37±0.3</td>
<td>33</td>
</tr>
<tr>
<td>South Asia(SAS)</td>
<td>59±6</td>
<td>57±0.8</td>
<td>44</td>
</tr>
<tr>
<td>Northern South America( NSA)</td>
<td>73±9</td>
<td>69±4</td>
<td>56</td>
</tr>
<tr>
<td>Southern South America(SSA)</td>
<td>33±4</td>
<td>31±2</td>
<td>27</td>
</tr>
<tr>
<td>Africa(AF)</td>
<td>76±4</td>
<td>74±1</td>
<td>65</td>
</tr>
</tbody>
</table>


<table>
<thead>
<tr>
<th>Model</th>
<th>90-30°S</th>
<th>30°S-0°</th>
<th>0°-30°N</th>
<th>30°-90°N</th>
<th>Global</th>
</tr>
</thead>
<tbody>
<tr>
<td>TransCom</td>
<td>0.0%</td>
<td>0.0%</td>
<td>0.0%</td>
<td>0.0%</td>
<td>0.0%</td>
</tr>
<tr>
<td>INCA NMHC</td>
<td>-0.5%</td>
<td>-0.9%</td>
<td>-0.3%</td>
<td>-0.2%</td>
<td>-0.5%</td>
</tr>
<tr>
<td>CESM1-WACCM</td>
<td>1.1%</td>
<td>1.6%</td>
<td>2.5%</td>
<td>1.2%</td>
<td>1.8%</td>
</tr>
<tr>
<td>EMAC-L90MA</td>
<td>-0.1%</td>
<td>0.1%</td>
<td>1.3%</td>
<td>1.1%</td>
<td>0.7%</td>
</tr>
<tr>
<td>GEOSCCM</td>
<td>-0.3%</td>
<td>1.1%</td>
<td>1.4%</td>
<td>1.0%</td>
<td>1.0%</td>
</tr>
<tr>
<td>MRI-ESM1r1</td>
<td>-2.0%</td>
<td>0.2%</td>
<td>2.4%</td>
<td>1.7%</td>
<td>1.1%</td>
</tr>
</tbody>
</table>

\(^a\) We do not analyze the hemispheric CH$_4$ emission estimated with MOCAGE and SOCOL3 OH field since inversions using the two OH fields calculate much higher CH$_4$ emissions than using other OH fields.
Table 5. Global total emission changes (in Tg yr\(^{-1}\)) from the early 2000s (2000/07/01-2002/06/01) to the late 2000s (2007/07/01-2009/06/01) calculated to identify the effect of OH fields (\(E_{\text{change\_all}}\), Inv3\(\rightarrow\)Inv2), of OH fixed to early 2000s (\(E_{\text{change\_fixoh}}\), Inv4\(\rightarrow\)Inv2), and the contribution of OH changes from the early to late 2000s to top-down estimated CH\(_4\) emissions changes (\(E_{\text{change\_varoh}}\), Inv4\(\rightarrow\)Inv3).

<table>
<thead>
<tr>
<th>Model</th>
<th>(E_{\text{change_all}}) Inv3(\rightarrow)Inv2</th>
<th>(E_{\text{change_fixoh}}) Inv4(\rightarrow)Inv2</th>
<th>(E_{\text{change_varoh}}) Inv4(\rightarrow)Inv3</th>
</tr>
</thead>
<tbody>
<tr>
<td>TransCom</td>
<td>17.2</td>
<td>17.2</td>
<td>0.0</td>
</tr>
<tr>
<td>INCA NMHC</td>
<td>16.6</td>
<td>19.3</td>
<td>-2.7</td>
</tr>
<tr>
<td>CESM1-WACCM</td>
<td>30.0</td>
<td>18.5</td>
<td>11.5</td>
</tr>
<tr>
<td>EMAC-L90MA</td>
<td>20.4</td>
<td>15.3</td>
<td>5.1</td>
</tr>
<tr>
<td>GEOSCCM</td>
<td>19.1</td>
<td>16.1</td>
<td>3.0</td>
</tr>
<tr>
<td>MRI-ESM1r1</td>
<td>27.8</td>
<td>14.3</td>
<td>13.5</td>
</tr>
<tr>
<td>Mean ± SD</td>
<td>21.9±5.7</td>
<td>16.9±1.9</td>
<td>5.1±6.4</td>
</tr>
</tbody>
</table>

Table 6. Global sectoral emission changes (in Tg yr\(^{-1}\)) from the early 2000s (2000/07/01-2002/06/01) to the late 2000s (2007/07/01-2009/06/01) (mean±SD).

<table>
<thead>
<tr>
<th>Sector</th>
<th>(E_{\text{change_all}}) Inv3(\rightarrow)Inv2</th>
<th>(E_{\text{change_fixoh}}) Inv4(\rightarrow)Inv2</th>
<th>(E_{\text{change_varoh}}) Inv4(\rightarrow)Inv3</th>
<th>Prior</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wetland</td>
<td>-3.5±2.5</td>
<td>-5.6±1.3</td>
<td>2.1±3.4</td>
<td>0.0</td>
</tr>
<tr>
<td>Agri-waste</td>
<td>14.2±2.1</td>
<td>12.3±0.7</td>
<td>1.9±2.3</td>
<td>19.0</td>
</tr>
<tr>
<td>Fossil fuel</td>
<td>8.7±0.8</td>
<td>8.1±0.8</td>
<td>0.6±0.9</td>
<td>18.0</td>
</tr>
<tr>
<td>Other</td>
<td>2.4±0.5</td>
<td>2.0±0.2</td>
<td>0.5±0.5</td>
<td>2.5</td>
</tr>
<tr>
<td>Total</td>
<td>21.9±5.7</td>
<td>16.8±1.9</td>
<td>5.1±6.4</td>
<td>39.4</td>
</tr>
</tbody>
</table>
Figures

Figure 1. A diagram showing inversion experiments (Inv1–Inv6) performed in this study. For each experiment, “Inv” gives the time period of inversion, and “OH” gives the time period of the OH fields used in the inversion. Inv1 is performed using the original OH field, whereas Inv2–Inv5 are performed using scaled OH fields. The colored boxes on the left and right show analyses of inversions we did to examine the OH impacts on inverted CH₄ emissions. The brown, yellow, and green textboxes correspond to analyses presented in Section 3.1, Section 3.2, and Section 3.3, respectively.
Figure 2. Left: The global CH$_4$ emissions from Inv1 (averaged over 2000/07/01-2002/06/01). The bottom-up and top-down estimations over 2000-2009 from previous GCP (Kirschke et al., 2013; Saunois et al., 2016) are also presented for comparison. The brown box plot shows the inversion results using 10 OH fields, while the orange one shows the results that exclude the largest estimates from the SOCOL3 and MOCAGE OH fields. The left, middle and right whisker chart (vertical lines) represent the minimum, mean, and maximum values of different inversion, and the left and right edges of boxes represent the mean ± one standard deviation. The definition of the box plot is applicable to all those hereafter. Right: The relationship between the optimized CH$_4$ emissions (Tg yr$^{-1}$) in Inv1 and the corresponding [OH]$_{GM-CH_4}$ ($\times 10^5$ mole cm$^{-3}$). The correlation coefficient (r) and the linear regression equation fitted to the data are shown inset.
Figure 3. Zonal (left), and regional averages (right) of CH\(_4\) emissions calculated by Inv1 (top row) and Inv2 (bottom row) with 8 OH fields from 2000/07/01 to 2002/06/01. Left and right panels: prior (dash red line) and mean optimized (solid red line) CH\(_4\) emissions for every 15-degree latitudinal band and 10 regions, respectively. Where USA=North America, CAN=Canada, EU=Europe, RUS=Russia, CHN=China, SEAS=Southeast Asia, SAS=South Asia, NSA=northern South America, SSA=southern South America, AF=Africa. The full names of the abbreviations are applicable to all figures hereafter. The differences between prior and optimized emissions (optimized minus prior) are shown by the box plots (defined in Fig. 1). Prior and optimized emissions values correspond to the right axes and their differences correspond to the left axes. Middle panel: the ratio of the standard deviation of emissions in each grid-cell calculated by Inv1 (top) and Inv2 (bottom) to prior emissions.
Figure 4. Global total CH$_4$ emissions from four broad categories from 2000/07/01-2002/06/01 in Tg yr$^{-1}$. The red circles and dots show the prior emissions and mean optimized emissions, respectively, as calculated by Inv1 (right axes), and the box plots (defined in Fig. 1) show the difference between optimized emissions calculated by Inv1 (blue) and Inv2 (yellow) and prior emissions (optimized minus prior, left axes).
Figure 5. Same as Fig. 4 but for prior and optimized emissions over 10 emitting regions covering most of the emitting lands. W=Wetlands, A=Agri-waste, F=Fossil fuels, and O=Others. In Tg yr$^{-1}$. 

995
Figure 6. Top panel: latitudinal emission (every 15-degree latitudinal band) changes in Tg yr$^{-1}$ from the early 2000s (2000/07/01-2002/06/01) to the late 2000s (2007/07/01-2009/06/01) of total, wetlands, agriculture and waste, and fossil fuel emissions (E_change_all, Inv3 – Inv2). Bottom panel: contribution of OH changes on top-down estimated CH$_4$ emission changes between the two periods (E_change_varoh, Inv3 – Inv4). The red lines are changes of prior emissions and the black lines are the mean changes of optimized emissions. The box plots (defined by Fig. 1) show the uncertainties and ranges calculated with different OH fields.
Figure 7. Top panel: optimized global total and sectoral regional emission changes in Tg yr$^{-1}$ from the early 2000s to the late 2000s (y-axis) plotted against prior emission changes between the two time periods (x-axis) as derived from Inv3 and Inv4(E_change_all). The prior wetland emissions are constant over time, thus show zero changes (all ‘0’ on the x-axis). Bottom panel: contribution of OH changes between the two periods on top-down estimated emission changes (E_change_varoh). The box plots (defined by Fig. 1) show the uncertainties and ranges calculated with different OH fields.
Figure 8. Top panel: Time series of global total CH$_4$ emissions calculated by Inv5 (yellow) and Inv6 (purple) plotted together with prior emissions (black), and [OH]$_{GM}$-CH$_4$ anomaly of CESM OH fields (blue). Bottom panel: the difference of global total (black line) and latitudinal (stack bar plots) CH$_4$ emissions between Inv5 and Inv6 (Inv5-Inv6).

Figure 9. Comparison between Inv5 (x-axis) and Inv6 (y-axis) estimated global total CH$_4$ emissions trends in Tg yr$^{-2}$ between 2004 and 2015 for the four categories (left) and over the ten continental regions (right). The error bars show the trend with 95% confidence intervals.